RKKY interaction of magnetic moments in nanosized systems

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Nanosized spherical system of magnetic moments interacting indirectly via the RKKY mechanism is studied. The interaction energy that determines the temperature of the ferromagnetic ordering, depends strongly on the system size. Obtained in the mean-field approximation, dimensional and concentration dependencies of the Curie temperature testify to the necessity of taking into account the finite size of such systems to calculate their features. Results may concern both artificially constructed nanosystems and naturally arising formations (such as clusters of magnetic ions in diluted magnetic semiconductors, etc.).

In systems with free carriers of high concentration (metals or degenerate semiconductors), indirect magnetic impurities' interaction of Ruderman-Kittel-Kasuya-Yosida (RKKY) type is considered as one of the basic mechanisms of the magnetic ordering [1]. There are a lot of papers dealing with RKKY interaction in three-, two- and one-dimensional systems of *infinite* size [1, 2, 3, 4]. However, to our knowledge, nobody considered how that interaction should be modified for systems of the *finite* size. In the present paper, we consider this problem for the case of the spherical system of the finite radius.

Estimating the energy $w(r) = -J(r)\mathbf{S}_1\mathbf{S}_2$ of the indirect RKKY-interaction of magnetic impurities with spins $\mathbf{S}_1$, $\mathbf{S}_2$ spaced at the distance $\mathbf{r}$ is based on making use of the expression

$$J(r) = \frac{J_{pd}^2}{N^2}\exp(-r/l){\sum_{\mathbf{q}}}'\sum_{\mathbf{k}} e^{i\mathbf{qr}}\frac{f(E_{\mathbf{k}})}{E_{\mathbf{k+q}}-E_{\mathbf{k}}}, \tag{1}$$

obtained in the second order of the perturbation theory [1]. Here, N is the number of lattice sites, J_{pd} is the exchange energy for the interaction of the impurity spin with a free charge carrier, $E_{\mathbf{k}}$ is the carrier energy, $f(E_{\mathbf{k}})$ is the Fermi-Dirac function which in the degenerate case equals $f(E_{\mathbf{k}}) = 1$ at $k < k_F$ and $f(E_{\mathbf{k}}) = 0$ at $k < k_F$, where k_F is the Fermi momentum. The prime by the the sum over q means that $q \neq 0$. The exponent $e^{-r/l}$ in (1) reflects the finite carrier mean free path l.

In the continual approximation, the summation in (1) is replaced by the integration which performed usually over *all* $k < k_F$ and $|\mathbf{k}+\mathbf{q}| > \mathbf{k_F}$. Then, the standard result corresponding to the case of the infinite system reads [1]:

$$J(r) = -I_0\Phi(r)\exp(-r/l), \quad \Phi(r) = \left(\frac{a}{r}\right)^4[\varphi(r)\cos\varphi(r) - \sin\varphi(r)], \tag{2}$$

where

$$I_0 = \frac{1}{32\pi^3}\left(\frac{ma^2}{\hbar^2}J_{pd}^2\right), \quad \varphi(r) = 2k_{F0}r, \tag{3}$$

a is the lattice constant, $k_{F0} = (3\pi^2 p)^{1/3}$ is the Fermi momentum of carriers of the concentration p.

In the case of finite system sizes or magnetic ions' clustering, the classic expression (2) for the energy of the RKKY-interaction should be rectified. For simplicity, we consider the case when magnetic ions form the spherical cluster. Due to the quasi-neutrality, its radius R determines not only the area where ions are arranged but also the region where carriers, produced by those ions, are localized. In other words, the carriers are contained in the potential well of the radius R. Therefore, the carrier momentum k and its variation q are limited by the intervals

$$k_1 \leq k \leq k_F, \quad k_1 \leq q \leq k_2, \tag{4}$$

where

$$k_1 \approx \pi/R,\ k_2 \approx \pi/a. \tag{5}$$

In addition, due to the spatial quantization the distance between energy levels of carriers grows that leads to increasing the Fermi energy and Fermi momentum with decreasing the well size: $k_F = k_F(R)$. Together, it complicates calculations and the final expression for $J(r)$ turns out to be more bulky than the canonical expression (2).

The finite mean free path l results in smearing energy levels of carriers due to their collisions. Therefore, the lowest value k is defined by the system size and equals $k_1 \approx \pi/R$ only if the collision broadening $\hbar/\tau$ of levels is less then the energy $\pi^2\hbar^2/2mR^2$ of the first level. That condition could be written in the form

$$\frac{\pi^2}{R^2} - \frac{2k_F}{l} > 0 \tag{6}$$

meaning that our approach relates to the small enough systems only. If, for instance, $l/a = 10$, $ak_F = 1$ then $R \lesssim 10a$. In the general case, one could use the value

$$k_1 = \sqrt{\max\left\{\left(\frac{\pi^2}{R^2} - \frac{2k_F}{l}\right), 0\right\}} \tag{7}$$

as the left boundary of inequalities (4).

To proceed one should estimate how the Fermi momentum depends on the cluster size. The total number of free carriers in the cube of the size R with the spherical Fermi surface is defined by the number of cells of the volume $(2\pi)^3$ in the phase space and in the limit of $k_F R \to \infty$ equals $N_F \sim k_F^3 R^3/(2\pi)^3$. For a finite $k_F R$-value, the number of carriers is defined by the number of points in the wavenumber space with coordinates divisible by $(2\pi/R)$. As none of those coordinates could equal zero, in that case

$$N_F \sim (k_F R/2\pi)^3 - \alpha(k_F R/2\pi)^2,$$

where the correction (proportional to $\alpha \sim 1$) is associated with the "forbidden" points, positioned in the coordinate planes of the wavenumber space and with the "correct" points neighboring to the Fermi surface but not fallen inwards, as well. For the carrier concentration $n = N_F/R^3$, it follows $n \sim k_F^3(1 - \alpha/k_F R)$. For $n = \text{const}$ and $k_F R \gg 1$ that leads to

$$k_F(R) - k_{F0} \approx \frac{\alpha}{3R},$$

where $k_{F0} \equiv k_F(\infty)$ is the Fermi momentum in the infinitely large system. Everywhere below, we use k_F to mean the value

$$k_F(R) = k_{F0} + \frac{1}{R}. \tag{8}$$

Let us turn now to calculating the energy of the RKKY interaction. Assuming $E_{\mathbf{k}} = \hbar^2 k^2/2m$, $E_{\mathbf{k+q}} = \hbar^2(k+q)^2/2m$ and designating the angle between $\mathbf{r}$ and $\mathbf{q}$ as α, and the angle between $\mathbf{k}$ and $\mathbf{q}$ as θ, we obtain

$$J(r) = \frac{2mJ_{pd}^2}{\hbar^2 N^2} \exp(-r/l)\, a^4 \sum_q{}' \frac{e^{iqr\cos\alpha}}{q} \sum_k \frac{f(E_k)}{2k\cos\theta + q}, \tag{9}$$

or, in the continual approximation,

$$J(r) = \frac{4}{\pi} I_0 \exp(-r/l)\, a^4 \int_{k_1}^{k_2} qdq \int_0^{\pi} e^{iqr\cos\alpha} \sin\alpha d\alpha \int_{k_1}^{k_F} k^2 dk \int_0^{\pi} \frac{\sin\theta d\theta}{2k\cos\theta + q}. \tag{10}$$

Non-complicated but laborious calculations lead to rather cumbersome result which could be represented in the relatively simple form by the help of the operator $\hat{L}$ determining the value of the double definite integral $\int_{k_1}^{k_F} dk \int_{k_1}^{k_2} (...) dq$ with the primitive function $\Psi(k,q)$:

$$\hat{L}\Psi(k,q) = \Psi(k_1,k_1) - \Psi(k_F,k_1) + \Psi(k_F,k_2) - \Psi(k_1,k_2).$$

Then

$$J(r) = \frac{1}{\pi} I_0 \exp(-r/l)\, (ak_F)^4 \hat{L}\Psi_r(k,q), \tag{11}$$

where

$$\Psi_r(k,q) = \frac{1}{(2k_F r)^4} \left[\Phi_1(k)\, c_1(k,q) - \Phi_2(k)\, c_2(k,q) + \Phi_3(k,q)\right], \tag{12}$$

$$\Phi_1(k) = 2kr\cos 2kr - \sin 2kr, \quad \Phi_2(k) = 2kr\sin 2kr + \cos 2kr, \tag{13}$$

$$c_1(k,q) = \mathrm{Si}[(2k+q)r] - \mathrm{Si}[(2k-q)r], \quad c_2(k,q) = \mathrm{Ci}[(2k+q)r] - \mathrm{Ci}(|2k-q|r), \tag{14}$$

$$\Phi_3(k,q) = \cos qr(1 + 2k^2r^2) \ln\left|\frac{2k+q}{2k-q}\right| + qr(\sin qr - \frac{1}{2} qr\cos qr)\left(\ln\left|\frac{2k+q}{2k-q}\right| - \frac{4k}{q}\right), \tag{15}$$

$$\mathrm{Si}(x) = \int_0^x \frac{\sin t}{t} dt, \quad \mathrm{Ci}(x) = -\int_x^{\infty} \frac{\cos t}{t} dt. \tag{16}$$

To the traditional situation ($R \to \infty$) there correspond $k_1 \to 0$, $k_2 \to \infty$. In that case $c_1 \to \pi$, $c_2 \to 0$, $\Phi_3 \to 0$, and

$$\hat{L}\Psi \to \frac{\pi}{(2k_F r)^4}\Phi_1(k_F) = \pi \frac{2k_F r\cos 2k_F r - \sin 2k_F r}{(2k_F r)^4}.$$

Hence, (11) reduces to the standard expression (2). For finite values k_1, k_2, the interaction energy $J(r)$ should be calculated with Eqs. (11)–(15).

The local effective RKKY-field H_{RKKY}, generated in a given point, is defined by the relation $\mu H_{\mathrm{RKKY}} = \sum_i J(r_i)$ where r_i is the distance from that point to the ith magnetic impurity, μ is

the impurity magnetic moment. In the continual approximation, the sum could be replaced by the integral $\mu H_{\mathrm{RKKY}} = \int J(r')d^3r'$ where the integration is spread over the volume occupied by impurities. Contrary to the infinite system, the value of that integral depends on the position of the considered point. For the spherical system, the effective field could be characterized by the value

$$\mu H^0_{\mathrm{RKKY}} = 4\pi \int_{r_{\min}}^{R} J(r)r^2 dr \tag{17}$$

of that integral in the center of the sphere. Here $r_{\min}$ is the minimum distance between impurities determined by the discreteness of the crystal lattice (for instance, the minimum distance between extrinsic Mn atoms, replacing Ga atoms in GaAs lattice, amounts $r_{\min} = a/\sqrt{2} \approx 4\,\text{Å}$).

How much would the local field H_{RKKY} be non-uniform inside the sphere one could judge by making note that in the case with the interaction length l being comparable or shorter than the radius R, the local field at the surface of the sphere should be approximately half as large as its value in the center of the sphere. It is not hard to show that for any point inside the sphere being offset by the distance $h \leq R$ from its center, one could employ the relation

$$\mu H_{\mathrm{RKKY}} = 4\pi \int_{r_{\min}}^{R+h} J(r)r^2 F(r) dr, \quad F(r) = \begin{cases} 1, & r < R-h \\ \dfrac{R^2 - (h-r)^2}{4rh}, & R-h < r < R+h \end{cases}, \tag{18}$$

instead of (17). In particular, for the field at the sphere surface ($h = R$) one could find

$$\mu H^S_{\mathrm{RKKY}} = 2\pi \int_{r_{\min}}^{2R} J(r)(1 - r/2R)r^2 dr. \tag{19}$$

The results of numerical calculations (see Fig. 1) with $r_{\min} = a/\sqrt{2}$, $l = 3a$, $k_{F0}a = 1$ show that for $R = 10a$ the part of the sphere where the effective field differs from H^0_{RKKY} no more than by 20%, amounts about 85% of its volume, and the average field $\bar{H}_{\mathrm{RKKY}} = (3/R^3) \int_0^R H_{\mathrm{RKKY}}(h)h^2 dh \approx 0.93 H^0_{\mathrm{RKKY}}$ (the same for $R = 3a$ amounts $\bar{H}_{\mathrm{RKKY}} \approx 1.14 H^0_{\mathrm{RKKY}}$). Hence, one could, in the first approximation, ignore the non-uniformity of the effective field and consider it as nearly uniform and equal to $\bar{H}_{\mathrm{RKKY}} \approx H^0_{\mathrm{RKKY}}$.

In the mean-field theory, Curie temperature T_{C} of the ferromagnetic state arising due to the RKKY-interaction is defined by the simple relation [1]

$$k_B T_{\mathrm{C}} \sim \mu \bar{H}_{\mathrm{RKKY}} \approx \mu H^0_{\mathrm{RKKY}}. \tag{20}$$

It is of interest to understand how the so-defined Curie temperature depends on the system size R at $k_{F0} = \mathrm{const}$ or varies with the carrier concentration (determined by the Fermi momentum [5]) in systems of fixed (but different) sizes [6]. Corresponding dependencies are shown in Fig. 2, 3.

The dependence $T_{\mathrm{C}}(R)$ turns out to be non-monotone, with pronounced oscillations of the period $2a$ at higher k_{F0} values. They are resulted from that part of the interaction energy $J(r)$ which is defined by the function $\Phi_3(k, q)$ given by (15), namely, by the terms $\sin qr$ and $\cos qr$ with $q = k_2 = \pi/a$. That provides for the observed period.

At $R \sim l$, the Curie temperature exceeds noticeably its value for $R \to \infty$ (shown by dot lines in Fig. 2). But the most significant distinction of the dependence $T_{\mathrm{C}}(R)$ (associated with accounting the finite k_1 value) is observed at small R. As soon as the condition (6) is reached, the drop of T_{C} is observed, as compared to the "standard" (corresponding to $k_1 = 0$) dependence (shown by dash curves in Fig. 2). That means the impossibility of existing ferromagnetism due to RKKY interaction in small clusters.

As for the dependence $T_{\mathrm{C}}(k_{F0})$, the Curie temperature increases almost monotonously with k_{F0} (i.e., with rising the carrier concentration). In that case also, reaching the condition (6) results in the considerable decreasing T_{C} (down to zero at some finite size $R \sim l$) as compared to the $T_{\mathrm{C}}(k_{F0})$ dependence for $R \to \infty$.

In conclusion, we studied nanosized spherical systems of magnetic moments interacting indirectly via the RKKY mechanism. The interaction energy which determines the temperature T_{C} of ferromagnetic ordering, depends strongly on the system size. Obtained in the mean-field approximation, dimensional and concentration dependencies of the Curie temperature testify to the necessity of taking into account the finite size of such systems to calculate their features. Results may concern both artificially constructed nanosystems and naturally arising formations (such as clusters of magnetic ions in diluted magnetic semiconductors [7, 8], etc.).

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References

[1] D.C. Matiss, *The theory of magnetism* (Harper & Row Publ., New York, Evanston, London, 1965).

[2] Y. Yafet, Phys. Rev. B. **36**, 3948 (1987) – 1D-system.

[3] B. Fisher and M. Klein, Phys. Rev. B. **11**, 2025 (1975) – 2D-system.

[4] D. N. Aristov, Phys. Rev. B. **55**, 8064 (1997) – 1D+2D+3D-systems.

[5] If carriers are delivered by the magnetic impurities placed in the points of the face-centered cubic lattice, their Fermi momentum equals $k_{F0}a = 2\pi(3pa^3/16\pi)^{1/3}$. For the diluted magnetic semiconductor $Ga_{1-x}Mn_xAs$, $k_{F0}a \approx 4.91(\gamma x)^{1/3}$ where $\gamma < 1$ is the fraction of those Mn-ions that substitute Ga-atoms and turn into acceptors. Herefrom, it follows, for instance, $k_{F0} = 1$, 1.5, 2 and 2.5 for $\gamma x \approx 0.01$, 0.03, 0.07 and 0.13, correspondingly ($p = 4\gamma x/a^3 = 2.16 \cdot 10^{22}\gamma x$ cm^{-3}).

[6] Another way to express $T_{\rm C}$ in terms of $H_{\rm RKKY}$ is to relate it to the minimum effective field (at the sphere surface): $k_B T_{\rm C} \sim \mu H^S_{\rm RKKY}$. The Curie temperature so defined is half as large as that shown in Fig. 2, 3.

[7] H. Raebiger, A. Ayuela and J. von Boehm, Phys. Rev. B **72**, 014465 (2005)

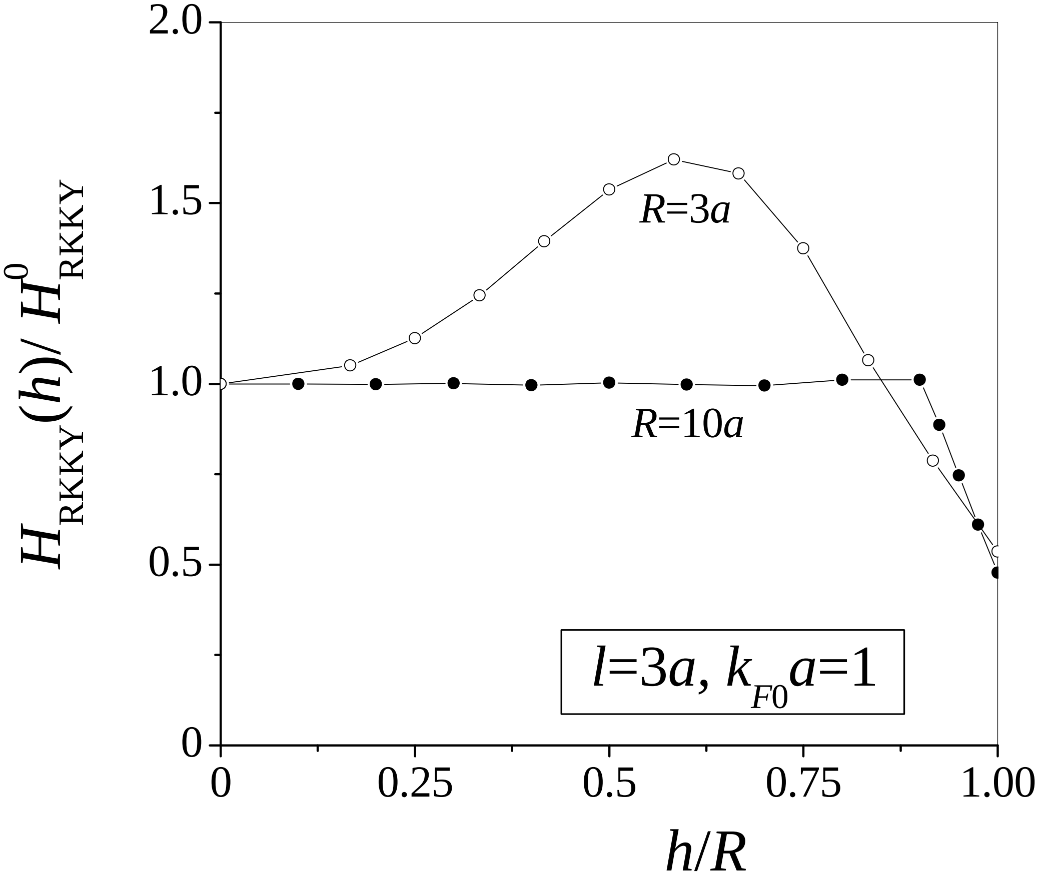
[8] D. J. Priour Jr. and S. Das Sarma, Phys. Rev. B **73**, 165203 (2006).

Captions

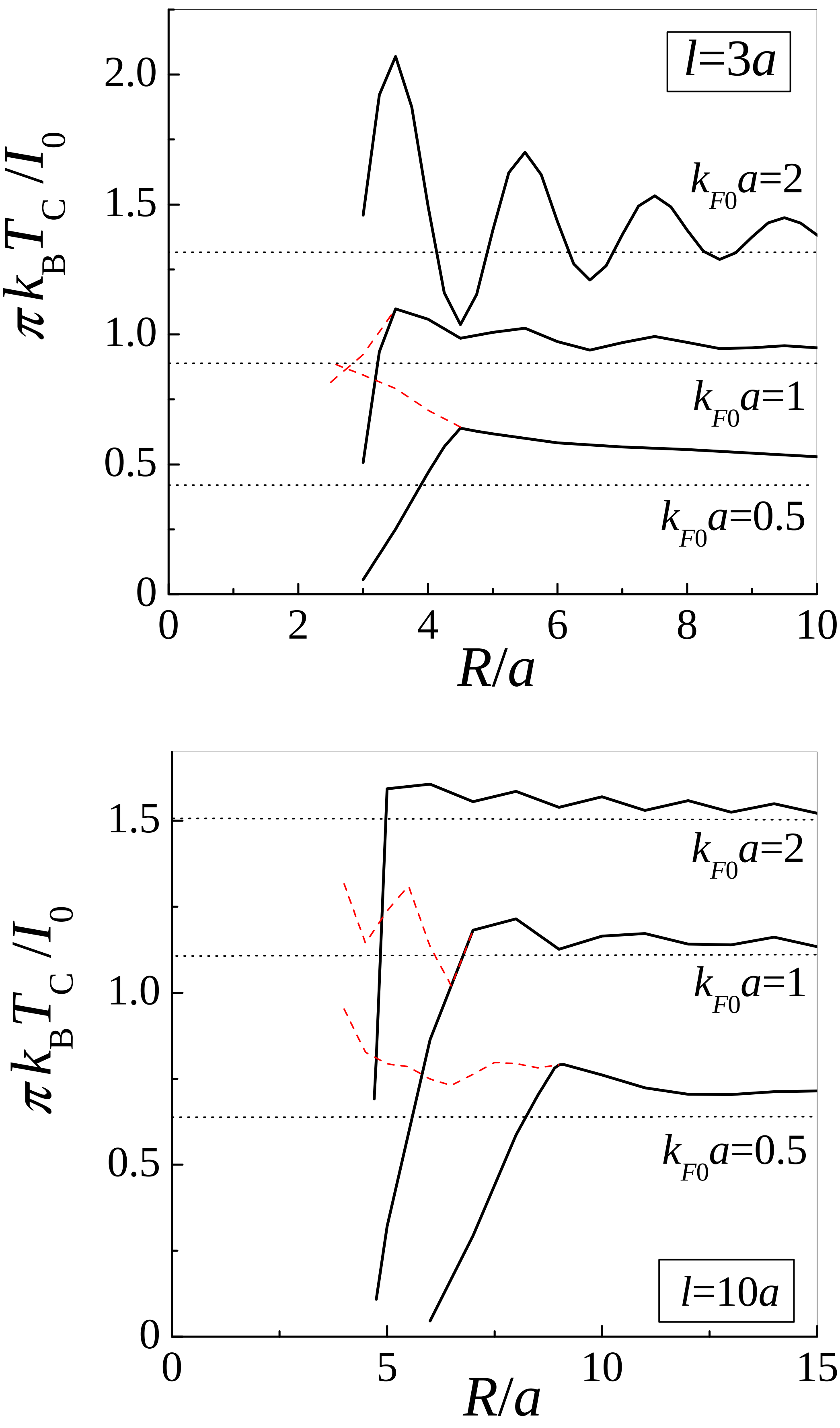
Fig. 1. Spatial distribution of the local effective field H_{RKKY} within the sphere of the radius $R = 10a$ and $R = 3a$ at $l = 3a$, $k_{F0}a = 1$.

Fig.2. Dependencies $T_{\mathrm{C}}(R)$ of the Curie temperature on the radius of spherical systems with various carrier concentrations. Upper panel: $l = 3a$, lower panel: $l = 10a$. Dot lines indicate the values $T_{\mathrm{C}}(R = \infty)$, dash curves show the "standard" (corresponding to $k_1 = 0$) behavior of the dependencies $T_{\mathrm{C}}(R)$ at small R.

Fig. 3. Dependencies $T_{\mathrm{C}}(k_{F0})$ of the Curie temperature on the carrier Fermi momentum for spherical systems of various radii. Upper panel: $l = 3a$, lower panel: $l = 10a$. Arrows indicate where the condition (6) is reached.



2.0
1.5
1.0
0.5
0
0
0.25
0.5
0.75
1.00
$H_{\mathrm{RKKY}}(h)/H^{0}_{\mathrm{RKKY}}$
h/R
R=3a
R=10a
l=3a, $k_{F0}a$=1



l=3a
k_{F0}a=2
k_{F0}a=1
k_{F0}a=0.5
πk_B T_C /I_0
R/a
0
0.5
1.0
1.5
2.0
2
4
6
8
10
k_{F0}a=2
k_{F0}a=1
k_{F0}a=0.5
l=10a
πk_B T_C /I_0
R/a
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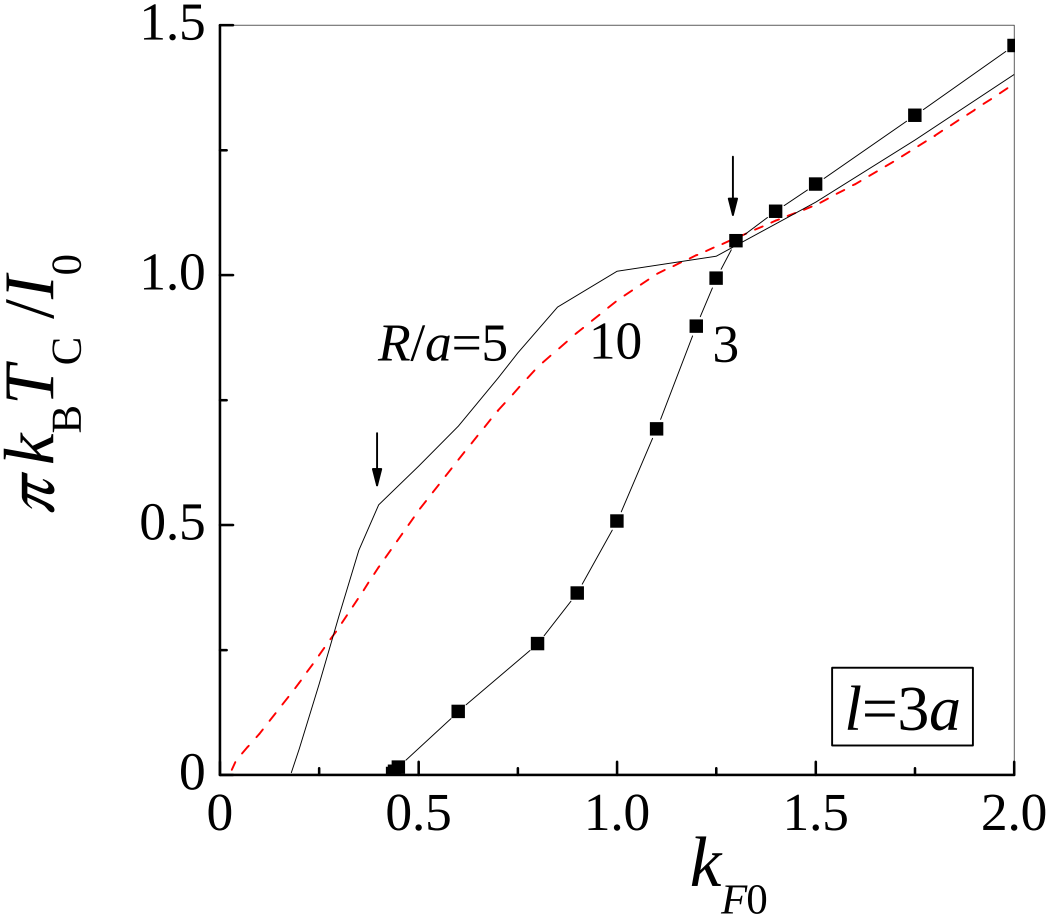
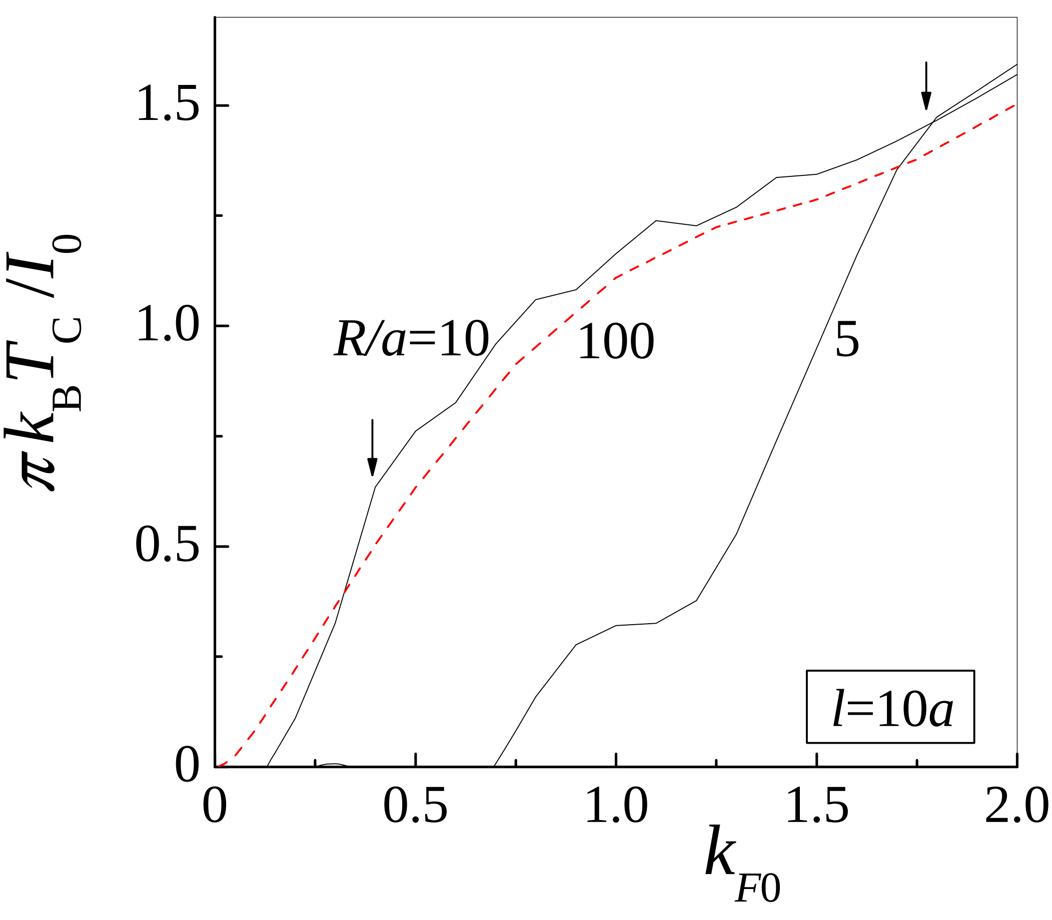
0.5
1.0
1.5
5
10
15



1.5
1.0
0.5
0
0
0.5
1.0
1.5
2.0
$\pi k_B T_C / I_0$
k_{F0}
R/a=5
10
3
l=3a



1.5
1.0
0.5
0
0
0.5
1.0
1.5
2.0
$\pi k_B T_C / I_0$
k_{F0}
R/a=10
100
5
l=10a